

Optical conductivity and the correlation strength of high temperature copper-oxide superconductors

Armin Comanac⁽¹⁾, Luca de' Medici⁽²⁾, Massimo Capone⁽³⁾, A. J. Millis⁽¹⁾

⁽¹⁾Department of Physics, Columbia University, 538 W. 120th Street, New York, NY 10027 USA

⁽²⁾Department of Physics and Center for Materials Theory, Rutgers the State University of NJ, 136 Frelinghuysen Road, Piscataway, NJ 08854

⁽³⁾SMC, CNR-INFM, and Dipartimento di Fisica, Universit di Roma "La Sapienza", Piazzale A. Moro 2, I-00185, Rome, Italy and ISC-CNR, Via dei Taurini 18, I-00185, Rome, Italy

High temperature copper-oxide-based superconductivity is obtained by adding carriers to insulating "parent compounds". It is widely believed the parent compounds are "Mott" insulators, in which the lack of conduction arises from anomalously strong electron-electron repulsion, and that the unusual properties of Mott insulators are responsible for high temperature superconductivity. This paper presents a comparison of optical conductivity measurements and theoretical calculations which challenges this belief. The analysis indicates that the correlation strength in the cuprates is not as strong as previously believed, that the materials are not properly regarded as Mott insulators, that antiferromagnetism is essential to obtain the insulating state and, by implication, that antiferromagnetism is essential to the properties of the doped metallic and superconducting state as well.

Since their discovery in 1986, the high temperature copper-oxide superconductors have been a central object of study in condensed matter physics. Their highly unusual proper-

ties are widely (although not universally) believed to be a consequence of electron-electron interactions which are so strong that the traditional paradigms of condensed matter physics do not apply: instead, entirely new concepts and techniques are required to describe the physics. In particular, the high- T_c materials are obtained by adding carriers to insulating parent compounds such as La_2CuO_4 . The lattice structure and electron counting of La_2CuO_4 is such that there is an odd number of electrons per formula unit. Thus, in the absence of further symmetry breaking, conventional band theory would predict that the material is a good metal. La_2CuO_4 is however not metallic; it is an insulator with a gap determined by optical spectroscopy to be approximately 1.8eV [1].

From one perspective the insulating behavior is not surprising. At temperature $T = 0$, La_2CuO_4 has two-sublattice Néel order, so that the *magnetic* unit cell contains two formula units and thus an even number of electrons, compatible with the observed insulating behavior. However, the general consensus has been that the antiferromagnetic order is irrelevant. Instead, the materials have been identified [2, 3] as 'Mott insulators': materials in which the electron-electron repulsion is so strong that the presence of an electron in one unit cell prevents another electron from entering that cell, independent of any electronic order. (While the cuprates are properly regarded as "charge-transfer" and not "Mott" insulators in the sense of Ref [4], this issue is not relevant here: the high energy scale physics and chemistry of transition metal (Cu) and ligand (O) ions produces one band of electrons, with an effective interaction strength which we aim to determine. In particular, optical data show that the nearest bands (arising in main from the non-bonding oxygen orbitals) are 5 – 6 eV removed in energy, with only a weak absorption tail extending down to the energies of relevance here. The issue is discussed in more detail in the supporting on-line material.) In a Mott or charge-transfer insulator, a density of one electron per unit cell implies a "jammed" situation: no electron can move without creating an energetically expensive doubly occupied site. Removing or adding electrons creates "holes" or doubly occupied sites, whose motion is not blocked by the jamming effect but is strongly affected by the nontrivial Mott insulating background in which it moves [3].

This paper argues that the experimental evidence is not in agreement with the strong correlation, "Mott" picture: rather, an intermediate coupling picture is appropriate, in which the antiferromagnetic order (or correlations) are crucial to the insulating behavior

and, by implication, to the physics of the doped, superconducting compounds. The important experimental evidence leading to this conclusion is the optical (frequency-dependent) conductivity, $\sigma(\omega)$; the linear response function connecting a frequency dependent, transverse electric field \mathbf{E} to the current \mathbf{j} it induces. At frequencies less than the interband threshold the measured conductivity is dominated by processes in which an electron moves from one unit cell to another. In a Mott insulator, such conductivity processes are suppressed by the blocking effect of on-site repulsion [5], so that the expected low frequency spectral weight (integrated optical absorption strength) is small. We show here that in the high- T_c materials the measured low energy spectral weight is too large to be compatible with the Mott (blocking) interpretation of the physics of the cuprates.

The electronic structure of the cuprates is such that one band (per CuO_2 unit) crosses the chemical potential; all other bands are full or empty and may to first approximation be neglected. Electrons moving in the relevant band are subject to an interaction whose most important component is a repulsion disfavoring configurations in which two electrons occupy the same site at the same time. This physics may be expressed mathematically via the "Hubbard" model of a band of electrons subject to local correlations. Although the Hubbard model is not a fully accurate description of the physics of high temperature superconductors, it contains the essence of the blocking effect and generally accepted [3] as the basic picture on which a more refined description should be based. We write the model in a mixed momentum (k) space position (i) space representation as

$$H = \sum_{k,\sigma} \varepsilon_k c_{k,\sigma}^\dagger c_{k\sigma} + U \sum_i \hat{n}_{i,\uparrow} \hat{n}_{i,\downarrow} \quad (1)$$

Here $\hat{n}_{i,\sigma}$ is the density operator for electrons of spin σ on site i and ε_k is the dispersion given by local density band calculations. Small variations among different calculations exist, but all agree within a few percent on the values of the parameters important for this study, which are the bandwidth $W \approx 3\text{eV}$ and the "kinetic energy" $K \approx 0.4\text{eV}$. For definiteness in this paper we use the ε_k derived from the "downfolding" parametrization of Ref. [6].).

At a density of one electron per cell the ground state of H , Eq 1 is believed to be a paramagnetic metal at small U (roughly $U < 1\text{eV}$) and an antiferromagnetic insulator at larger U , with a small range of antiferromagnetic metal in between. The key question

is whether the antiferromagnetic order is essential to the insulating nature of the ground state. To determine this we turn to the single-site dynamical mean field approximation [7]. In this approximation, spatial correlations between fluctuations are neglected but temporal fluctuations on a given site are included exactly. If long ranged antiferromagnetic order is not included in the calculation, one finds at carrier concentration $n = 1$ and temperature $T = 0$ a critical value $U_{c2} \approx 1.45W$ separating a small U metallic phase from a large U insulating phase. The band theory estimate $W \approx 3eV$ implies $U_{c2} \approx 4.4eV$. This large U phase is identified as a Mott insulator because an energy gap exists at the chemical potential in the absence of any intersite magnetic correlations.

We calculated the optical conductivity implied by Eq. 1, representing the electric field via a vector potential \mathbf{A} , using the minimal coupling $k \rightarrow k - A$ and standard linear response theory and multiplying the calculated result (a dimensionless conductance per CuO_2 plane) by the conductance quantum e^2/\hbar and dividing by the mean *LSCO* interplane distance $d = 6\text{\AA}$. The two main panels of Fig 1, which plot the calculated conductivity for several carrier concentrations at a value of U slightly greater than U_{c2} and one somewhat less. Consider the $x = 0$ results, representative of the parent compounds of the high- T_c materials. The $U > U_{c2}$ calculation reveals Mott insulating behavior: even if magnetic order is neglected the result is insulating (gap in the conductivity spectrum). Adding antiferromagnetism increases the gap and produces structure at the gap edge. On the other hand, the $U < U_{c2}$ calculation reveals metallic behavior (no gap) in the absence of antiferromagnetism while the antiferromagnetic calculation reveals a large gap.

To interpret the results we note that in models such as Eq 1 the optical conductivity obeys a "restricted f-sum rule" [5, 8]. Defining

$$K(\Omega) = \left(\frac{V_{cell}}{a^2} \right) \int_0^\Omega \frac{2d\omega}{\pi} \frac{\sigma(\omega)}{\sigma_Q} \quad (2)$$

we have

$$K(\infty) = \sum_{k,\sigma} n_{k,\sigma} \frac{\partial^2 \varepsilon_k}{\partial k_x^2} \quad (3)$$

Here $\sigma_Q = e^2/\hbar$ is the conductance quantum, V_{cell} is the volume of the unit cell, a is the in-plane lattice constant and $n_{k,\sigma}$ is the probability that the state of momentum k and spin σ is occupied. Note that $\sigma(\omega)$ in Eq 2 refers to the real (dissipative) conductivity calculated from

Eq 1; in physical terms it corresponds to that contribution to the measured conductivity arising from transitions within the band of states described by Eq 1. In a real material, interband transitions not described by Eq 1 also contribute to the conductivity; these make up the difference between Eq 3 and the familiar f-sum rule $\int_0^\infty d\omega \sigma(\omega) = \pi n e^2 / 2m$.

If $U = 0$, $n(k, \sigma)$ is the usual Fermi-Dirac distribution, corresponding to filling up only the lowest-lying states in the band. Evaluation of Eq 3 for this case yields $K = K_{band} \approx 0.4eV$, essentially independent of carrier concentration for the dopings relevant to high temperature superconductivity. In this noninteracting case the electrons are not scattered; they are freely accelerated by an applied electric field so the conductivity is just a delta function of strength πK at $\omega = 0$. Increasing the interaction causes electron-electron scattering which shifts spectral weight from $\omega = 0$ to higher frequencies. Increasing the interaction also tends to localize the electrons, leading to an $n(k)$ more uniformly distributed over the band and thus reducing the magnitude of the integral in Eq 3, i.e. decreasing the total spectral weight. However, adding holes allows carrier motion, thus increasing the spectral weight and shifting it back towards $\omega = 0$. These effects can be seen in the insets of Fig. 1 which plot the conductivity integral, Eq 2 obtained from the calculated conductivities shown in the main panel of the figure.

We now compare the calculation to measurements of the conductivity, of which a representative example [9] is shown in Fig. 2. These data were taken in 1991; subsequent improvements especially in sample quality have sharpened the band gap seen in the conductivity of the $x = 0$ sample, so that the onset of absorption begins at $\omega \approx 1.8eV$ but have not changed the material features; in particular the spectral weights in the different frequency regimes. Use of the band theory estimate $W \approx 3eV$ would imply the band gap is approximately $0.6W$, consistent with the result of the antiferromagnetic-phase $U < U_{c2}$ calculation but inconsistent with the antiferromagnetic phase $U > U_{c2}$ result. The antiferromagnetic $U > U_{c2}$ calculation can be made consistent with the observed band gap by reducing the energy scales by 25%, implying in particular a bandwidth $W^* \approx 2.25eV$ instead of the $W \approx 3eV$ found in band theory calculations. However, even if this renormalization is made, the magnitude of the observed conductivity is inconsistent with the $U > U_{c2}$ hypothesis, as will now be shown.

The measured spectral weight in the range $\omega < 3eV$ for the insulating compound corre-

sponds to $K(3eV) = 0.2eV$ or about 50% of the noninteracting value. It is likely that not all of the spectral weight observed in the range below $3eV$ is due to the optical transitions of interest. Interband transitions to irrelevant bands may contribute. To obtain an upper bound on possible interband contributions we note that as doping increases the calculated conductivity shifts strongly to lower frequencies (as may be seen in Fig. 1). We therefore use the measured $x = 0.34$ data in the range $\omega > 1eV$ as an estimate of the interband contribution to the conductivity. We have integrated the difference between the conductivity measured in the $x = 0$ sample and that measured in the $x = 0.34$ sample over the range $\omega < 3eV$ obtaining $K_{exp}(\Omega = 3eV) \approx 0.2K_{band} \approx 0.1eV$. This estimate is quite consistent with the results shown in the inset of the lower panel of Fig 1. However, the $U > U_{c2}$ calculation yields substantially less spectral weight in the low frequency regime. Combining the band theory estimate $W \approx 3eV$ with the data in the inset of the upper panel of Fig. 1 yields $K(3eV) \approx 0.03eV$, far less than the measured $0.1eV$. If we use instead the renormalized $W^* = 2.25eV$ which reproduces the value of the gap, then $3eV \approx 4W^*/3$. The total spectral weight integrated up to this point is $0.13K(U = 0)$ and because the theoretical $K(U = 0) \sim W$ this implies an integrated weight of about $0.05eV$ still much smaller than what is observed.

Now La_2CuO_4 is observed to remain insulating at temperatures above its Néel temperature $\approx 340K$, so *long ranged* order is not essential to the insulating behavior. However, the Néel temperature is strongly suppressed by low dimensional fluctuation effects and is a poor measure of the strength of the magnetic correlations, which are found to remain significant up to the highest measured temperatures ($T \approx 1000K$) [11]. Recent cluster dynamical mean field calculations (K. Haule et. al., private communication; E. Gull, P. Werner, M. Troyer and A. Millis, to be published) produce insulating behavior over wide temperature ranges without long ranged order, even in the intermediate coupling regime, provided that near-neighbor spin correlations are strong enough.

We next turn to the doping dependence of the conductivity. The solid symbols in Fig. 3. show the optical spectral weight for several cuprate materials, integrated up to $0.8eV$, about 0.45 of the insulating gap. The value is chosen because available evidence indicates that the conductivity at $\omega < 1eV$ is essentially uncontaminated by interband transitions while at higher frequencies the situation is less clear [10]. One sees that the

measured spectral weight in the mid-gap region scales linearly with doping, but with a non-vanishing intercept. The open symbols show the results of the theoretical calculations for a U slightly greater than U_{c2} and for two U -values less than U_{c2} . For the $U > U_{c2}$ calculation we have used the scale $W^* = 2.25eV$ to convert the theoretical results to physical units. We see that for $U > U_{c2}$ the calculated spectral weight is qualitatively inconsistent with the data, because it vanishes as doping tends to zero. However, we note that in the qualitative comparison the decisive feature is the behavior at $x < 0.1$ where the uncertainties in the data are largest. Further experimental examination of this frequency regime would be desirable. The $U > U_{c2}$ results are also somewhat smaller in magnitude than the experimentally determined values. On the other hand, the results for $U = 0.9U_{c2}$ give a magnitude and doping dependence which is reasonably consistent with the measured values at non-vanishing dopings. The $x > 0$ calculations are performed within single-site dynamical mean field theory in the paramagnetic phase. This method does not take into account the effects of near-neighbor magnetic correlations, which are likely to be present even in the absence of true long ranged order and which will suppress somewhat the spectral weight in the low frequency regime. We suggest that a model with a $U \approx 0.85U_{c2}$ and with a proper treatment of antiferromagnetic correlations will lead to a doping dependence of the spectral weight which is consistent with experiment.

The results presented here suggest that a re-examination of theoretical approaches to high temperature superconductivity would be worthwhile. Much work has been based on the "t-J" model [3], which is derived on the assumption that the correlation-induced blocking effect is fundamental, with antiferromagnetism providing a next correction and which has been widely accepted because it provides natural explanation of the striking doping dependence of physical properties. Determining whether the observed doping dependence of the low energy physics can be understood within the intermediate-coupling, strong antiferromagnetic correlations picture implied by the optical data, is an important challenge for future work.

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Figure Captions

Fig. 1: Optical conductivity of Hubbard model: calculated as described in main text at dopings $x = 1 - n$ and interaction strength U indicated. Main panels: optical conductivity; insets: optical integral. For $x = 0$ both paramagnetic and antiferromagnetic phase calculations are shown; for $x > 0$ only paramagnetic phase results are given. If the band theory value $W = 3eV$ is used then the frequency scale is electron volts.

Fig. 2: Measured optical conductivity of $La_{2-x}Cu_xCuO_4$ reproduced from Ref. [9]); solid vertical line at frequency $\omega = 0.8eV$ indicates cutoff frequencies used for spectral weight analysis.

Fig. 3: Comparison of measured and calculated optical spectral weight. Solid symbols: spectral weight obtained by integrating experimental conductivity up to $0.8eV$ from references given. Open symbols: theoretically calculated spectral weight, integrated up to $W/4$. For $U = 0.85U_{c2}$ and $U = 0.9U_{c2}$ the band-theory estimate $W = 3eV$ is used to convert the calculation to physical units; for $U = 1.02U_{c2}$ the value $W = 2.25eV$ which reproduces the insulating gap is used.

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- [1] D. Basov and T. Timusk, Rev. Mod. Phys. **77**, 721-80 (2005).
- [2] P. W. Anderson, Science, **235**, n 4793, 1196-86 (1987).
- [3] Patrick A. Lee, Naoto Nagaosa, and Xiao-Gang Wen, Rev. Mod. Phys. **78**, 17-86 (2006)
- [4] J. Zaanen, G. Sawatzky and J. Allen, Phys. Rev. Lett. **55** 418 (1985).
- [5] A. J. Millis, “Optical Conductivity and Correlated Electron Physics” in *Strong Interactions in Low Dimensions*, D. Baeriswyl and L. Degiorgi, eds., Springer Verlag, Berlin (2004).
- [6] O. K. Andersen, A. I. Liechtenstein, O. Jepsen, and F. Paulsen, *J. Phys. Chem. Solids* **56**, 1573 (1995).
- [7] A. Georges, G. Kotliar, W Krauth, M. J. Rozenberg, Rev. Mod. Phys. **68**, 13 (1996).
- [8] P. F. Maldague, Phys. Rev. **B16**, 2437 (1977).
- [9] S. Uchida, T. Ido, H. Takagi, T. Arima, Y. Tokura, S. Tajima, Phys. Rev. **B43**, 7942 (1991).
- [10] A. J. Millis, A. Zimmers, R. P. S. M. Lobo, N. Bontemps and C. C. Homes, Phys. Rev. **B72** 24517/1-13 (2005).
- [11] T. Imai, C. P. Slichter, K. Yoshimura, M. Katoh, and K. Kosuge Phys. Rev. Lett. **71**, 1254 (1993).
- [12] A. Lucarelli, S. Lupi, M. Ortolani, P. Calvani, P. Maselli, M. Capizzi, P. Giura, H. Eisaki, N. Kikugawa, T. Fujita, M. Fujita, and K. Yamada, Phys. Rev. Lett. **90**, 037002 (2003)
- [13] Y. Onose, Y. Taguchi, K. Ishizaka, and Y. Tokura, Phys. Rev. **B69**, 024504/1-13 (2004)
- [14] S. L. Cooper, D. Reznyk, A. Kotz, M.A. Karlow, R. Liu, M.V. Klein, W.C. Lee, J. Giapintzakis, D.M. Ginsberg, B. W. Veal and A. P. Paulikas Phys. Rev. **B47**, 8233 (1993).
- [15] J. Hwang, T. Timusk and G. D. Gu, J. Phys. Condens. Matter **19** 125208 (2007).

Fig. 1

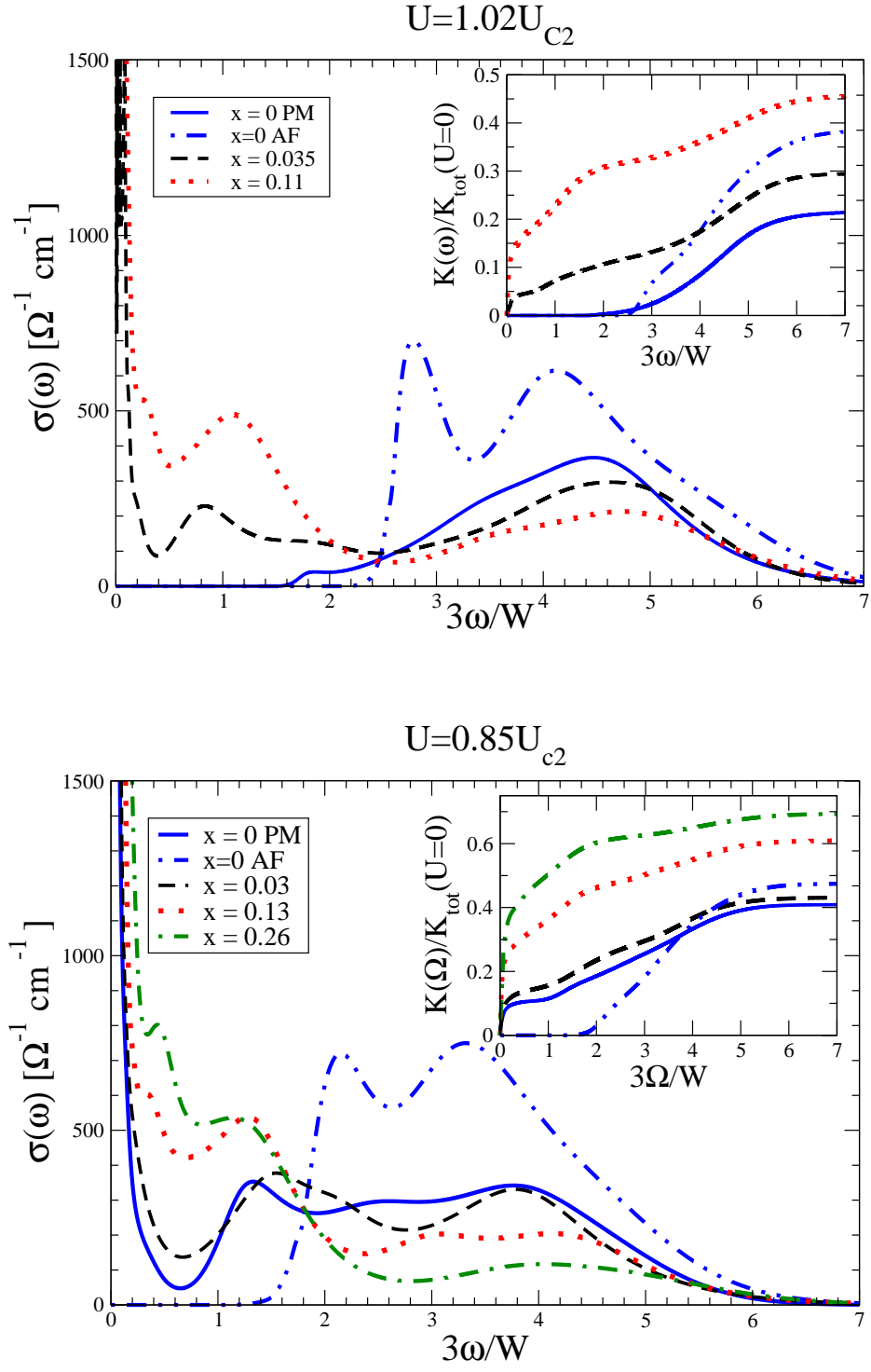


Fig. 2

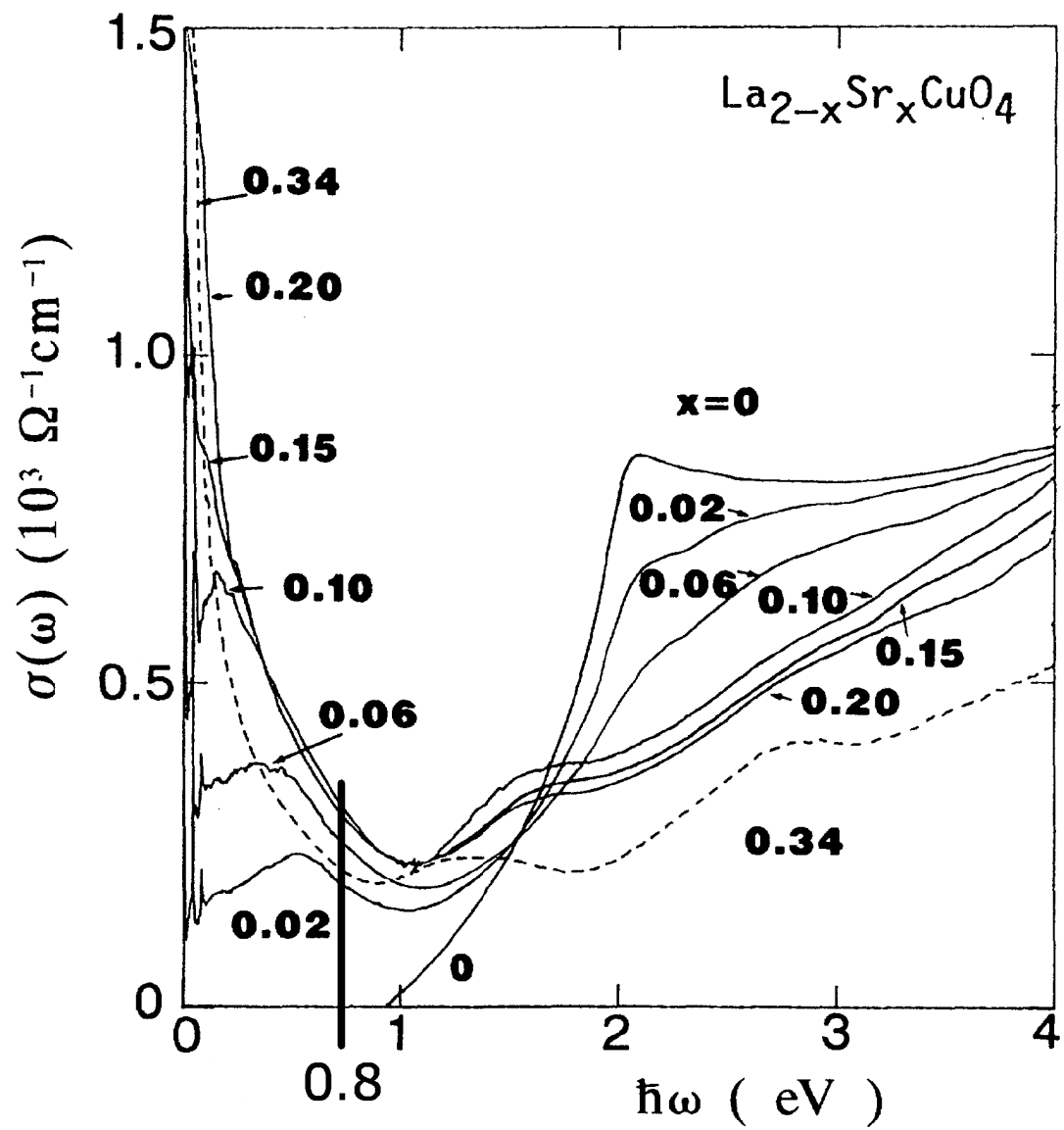


Fig. 3

